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FLAT MICROMETER-SIZED GRAPHITE, PROCESS FOR ITS PREPARATION, AND ITS  
APPLICATIONS

Applicant:  
(for all the contracting  
nations except U.S.A.):

CNRS  
National Center of  
Scientific Research  
15,, quai Anatole-France  
F-75700 Paris (FR).

Inventors (U.S. only):

Guy Furdin  
5, rue Paul-Eluard,  
Dommartin-sous-Amance,  
F-54770 Bruxières-aux-  
Chênes

Jean-Francois Mareche  
10, allée la Papellerie,  
F-54250 Champigneulles  
(FR)

Albert Herold  
6, rue E.-Albert, F-54520  
Laxou (FR)

Agent:

Yvette Sueur, etc.;  
Cabinet Sueur &  
L'Helgoualch, 78, rue  
Carnot, F-95240  
Cormeilles-en-Parisis  
(FR).

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micrometric graphite particles, their preparation method and use are described. This graphite material is characterized in that it essentially consists of single crystal particles with a particle-form factor of 50-1000, which have, when deposited in a layer a few millimetres thick on a plane support, a mosaic structure in which the angular carbon plane misalignment is  $\pm 15^\circ$ , as determined by X-ray diffraction. The flat micrometric graphite is useful as an electrode material in rechargeable or non-rechargeable batteries.

The present invention relates to micrometer-sized graphite particles, a process for their preparation, and to their applications.

Different powdered carbon-containing particles, graphitized or not, are known. They are industrially prepared by processes using different technologies.

The carbon blacks are obtained by the cracking of hydrocarbons in the vapor phase. They are present in the form of a nanometric powder whose particles are agglomerated or not. Certain of these materials can be graphitized by a later heat treatment.

The natural graphites are obtained after washing of the ore and purification of the graphite.

The artificial graphites are obtained by a controlled treatment of precursors (pyrolysis of coal or petroleum pitch), followed by graphitizing. The resulting products are then crushed dry in ball mills, or crushed wet in ball mills. Each product is referenced by its grain size. It is thus possible to distinguish the large flakes (diameter between 0.2 and 2 mm), the small flakes (diameter less than 0.35 mm), the fine particles (diameter less than 112  $\mu\text{m}$ ), and the very fine particles (diameter less than 20  $\mu\text{m}$ ).

For numerous applications of the lubrication type, one seeks to maintain the lamellar structure, that is to say an ultrathin

crystalline structure. The conventional crushing techniques break the grains in all directions.

An objective of the present invention is to provide a graphitized material in the form of flat micrometer-sized graphite particles.

Another objective of the invention is a process enabling one to obtain the flat micrometer-sized particles from carbon substances that can be graphitized or partially graphitized.

Finally, the invention relates to the applications of said material.

The graphitized material according to the present invention is characterized by the fact that it essentially consists of monocrystalline particles with a form factor between 50 and 1000, and which, after deposition on a flat support in the form of a layer a few millimeters thick, have a mosaic structure, determined by X-ray diffraction, on the order of a  $\pm 15^\circ$  angle.

Preferably, the form factor of the particles is greater than 100.

The form factor represents the ratio of the diameter  $\phi$  of the particles to their thickness  $d$ .

The graphitized material of the present invention is obtained by a two-step process. In a first step, one prepares an expanded material from a precursor material that can be or is graphitized. In a second step, the expanded material is subjected to crushing. The crushing results from the alternating action of an ultrasound probe and a rotating crusher with a shearing and cavitating effect. The alternation of these two types of crushing is particularly effective for fragmenting the precrushed expanded graphite particles into crystallite units. In effect, the combination of ultrasound and the rotating crusher enables one to obtain fine

particles with a very high form factor, on the order of 100 to 200.

The duration of the crushing sets the maximum size of the particles.

As an example of a rotating crusher with a shearing effect, it is possible to mention the Ultra Turrax crushers of the Janke and Kunkel company.

Preferably, the expanded material is subjected to precrushing for the essential purpose of wetting the expanded graphite with an appropriate liquid. Any liquid that wets the graphite can be used. As examples, it is possible to mention benzene, toluene, and cyclohexane, with benzene and toluene being particularly effective.

One preferably uses mixtures containing 1 to 60 g/L of expanded graphite. The mixture of expanded graphite and wetting liquid is precrushed, for example, by a helicoidal turbine. This precrushing stage makes homogenization of the mixture possible and leads to extensive fragmentation of the flakes of expanded graphite.

Drying after the crushing, to eliminate the wetting liquid, is useful for preventing agglomeration of the particles. The wetting liquid must be eliminated in order to avoid any recompaction of the particles of flat micrometer-sized graphite. This elimination can be advantageously done by lyophilization, preferably when benzene or cyclohexane is used as wetting liquid.

As precursor material, it is possible to use any substance that is or can be graphitized and in which an element can be inserted. As examples, it is possible to mention natural graphite of Madagascar or natural graphite of Brazil. Madagascar graphite is in the form of quasi-monocrystalline particles with an average diameter of 0.5 mm and an average thickness of a few tenths of a millimeter. It contains less than 0.5% ash. The Brazil graphite



is in the form of quasi-monocrystalline particles with an average diameter of 40  $\mu\text{m}$  and an average thickness of some tens of micrometers to a few hundred micrometers. It is also possible to mention the pitch cokes, which are or can be graphitized.

The precursor material can be expanded by a process consisting of intercalating a reagent in the precursor material, then subjecting the intercalated composite obtained, and possibly washed with water and dried, to a sudden temperature rise (thermal flash). The rapid consumption of the inserted material is accompanied by irreversible unidimensional expansion (or exfoliation) of each graphite particle. As the reagent, it is possible to use, for example, ferric chloride-ammonia mixtures or sulfuric acid. However, the reagents that undergo exothermic decomposition, such as those described in the patent application FR 91 97516, are particularly advantageous. Among these reagents that undergo exothermic decomposition, it is possible to mention perchloric acid, used alone or in a mixture with nitric acid, or nitromethane.

The apparent density of the expanded graphite thus obtained is very low (a few grams per  $\text{dm}^3$  as opposed to 2 kg per  $\text{dm}^3$  for the precursor material). Its specific [surface] area is on the order of a few tens of  $\text{m}^2/\text{g}$ , while it is on the order of 1  $\text{m}^2/\text{g}$  for the precursor material.

The present invention is described in more detail in the following examples, given on an illustrative but nonlimiting basis, and which relate to the preparation of the graphitized material of the present invention from different materials that are or can be graphitized.

The histogram of grain size distribution was established by counting and determining the size of the particles using an apparatus that applies light diffusion, for example, of the Malvern

type manufactured by Instrumat, which uses diffusion of a laser beam by the particles put in suspension in a carrier liquid. The histogram extends between 0 and 100  $\mu\text{m}$ . It depends on the time of crushing in the first four hours and on the wetting power of the liquid used. It gives the principal size of the particles.

The morphology of the product obtained is determined by examination with a scanning electron microscope. The particles are deposited on a previously polished brass support. The accelerating voltage of the incident beam of electrons is 21 kV. The thickness of the particles is such that the streaks of the support are visibly transparent, as are the nodules of copper in the brass matrix. This phenomenon, connected with the existence of retro-diffused electrons, reveals particles whose thickness is less than 0.1  $\mu\text{m}$ . This electron microscope examination gives point by point results.

The specific area of the flat micrometer-sized graphite obtained was evaluated using the isotherms of adsorption of krypton done at 77 K. The qualitative analysis of the isotherms shows that the product is characterized by a surface homogeneity identical to that encountered with a natural graphite. The quantitative use of the isotherms enables one to determine the specific area of the flat micrometer-sized graphite. Taking into account the lamellar morphology of the particles of flat micrometer-sized graphite, the covering power is deduced from this.

The crystallinity of the material is determined by electron diffraction. The particles are deposited by filtration on a nucleopore membrane before being recovered on an observation screen. The small thickness of the particles enables one to produce the diffraction conditions giving maximum resolution and

resulting in a single-point diffraction picture containing the [hko] reflections of the graphite.

The deposition ability of the flat micrometer-sized graphite particles was evaluated using a layer of particles deposited by settling on a beryllium plate with a small thickness. The X-ray diffractometric recording, done in transmission, shows the hko reflections, but also shows the 002 reflections, whose presence and intensity allow for suitable evaluation of the state of disorientation of the particles.

#### Example 1

A natural graphite, UF4, provided by Le Carbone Lorraine, and having a grain size distribution histogram compatible with an average diameter of 6.4  $\mu$ m, was used.

1 g of this material was added to 4 mL of a mixture composed of 70% commercial perchloric acid and fuming nitric acid (50/50 by volume) for 2 h. After reaction, this heterogeneous medium was filtered under vacuum. The cake recovered after filtration was placed in the bottom of a reactor heated beforehand to a red hot state. This first step led to the expansion of the particles of the starting material. The expanded particles were then subjected to precrushing for approximately 0.25 h using a helicoidal turbine at 2000 rpm. This was subjected to crushing for 3 h, with said crushing consisting of alternating sequences of 10 min of crushing with an Ultra-Turrax rotating crusher and sequences of 5 min of ultrasound. For this crushing step, the particles were wetted by cyclohexane.

The product obtained had a distribution histogram compatible with an average diameter of 6.2  $\mu$ m. Observation of the particles

with the scanning electron microscope exhibited a morphological state that was very different from the material before crushing. After treatment, the particles were transparent under the impact of the electron beam. The diameter/thickness ratio therefore greatly increased after crushing.

#### Example 2

The mode of operation of Example 1 was repeated with a non-graphitized pitch coke obtained by the carbonization of a coal pitch at 1100°C. This nongraphitized pitch coke had a histogram compatible with an average diameter of 50  $\mu$ .

The product obtained had a distribution histogram compatible with an average diameter of 28  $\mu$ .

#### Example 3

The mode of operation of Example 1 was repeated with a non-graphitized pitch coke, obtained by a heat treatment at 2500°C of the nongraphitized pitch coke used in Example 2. This graphitized pitch coke had a histogram compatible with an average diameter of 50  $\mu$ .

The product obtained had a distribution histogram compatible with an average diameter of 12  $\mu$ .

These results show that the process of the present invention is effective for cokes, and more particularly for graphitized cokes, which are therefore easily inserted.

#### Example 4

An intercalated composite was prepared from Madagascar graphite and sulfuric acid.

The intercalated composite obtained was washed with water and dried at 110°C. A sudden rise of the temperature to 1000°C caused the expansion of the graphite.

Different mixtures containing 1 to 60 g of expanded graphite in 1 L of benzene were subjected to precrushing for approximately 0.25 h using a helicoidal turbine at 2000 rpm. Each mixture was then alternately subjected to an ultrasound probe (20 kHz) and to a crusher of the Ultra Turrax type (20,000 rpm). Each sequence lasted 15 min, including 5 min of ultrasound and 10 min of Ultra-Turrax. The reactor was thermostatically controlled in order to avoid evaporation of the carrier liquid.

The benzene was eliminated by solidification of the mixture at 0°C and dynamic pumping of the vapors with the trapping of these at 180°C. After this last treatment, the flat micrometer-sized graphite was obtained in the dry state.

The distribution of the size of the particles subjected to stirring in a carrier liquid was determined using an apparatus of the Malvern type. Figure 1 gives the change in the size of the particles as a function of the crushing time for a mixture of 1 g/L of graphite. One notes that all the particles have a diameter less than 30  $\mu\text{m}$ .\*

The particles obtained have a thickness less than 0.1  $\mu\text{m}$ , a specific area on the order of 20  $\text{m}^2/\text{g}$ , and consequently a covering power of 10  $\text{m}^2/\text{g}$ .

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\* [Editor's note: Units used earlier were "mp."]

The electron diffraction diagram of the flat micrometer-sized graphite obtained indicates a quasi-perfect monocrystallinity of the material.

A layer a few tenths of a mm thick, which results from the superposing of several thousands of elementary particles, is characterized by a disorientation of the carbon-containing planes of a  $\pm 15^\circ$  angle. This result gives an account of the excellent ability of the material to be deposited flat.

#### Example 5

An intercalated composite was prepared from Brazil graphite and a mixture of nitric acid-perchloric acid-water according to the mode of operation described in the patent application FR 91 97516.

The intercalated composite obtained was washed with water and dried at  $110^\circ\text{C}$ . A sudden rise of the temperature to  $600^\circ\text{C}$  caused the expansion of the graphite.

Different mixtures containing 1 to 60 g of expanded graphite in 1 L of benzene were subjected to precrushing for approximately 0.25 h using a helicoidal turbine at 2000 rpm. Each mixture was then alternately subjected to an ultrasound probe (20 kHz) and to a crusher of the Ultra Turrax type (2,000 rpm). Each sequence lasted 15 min, including 5 min of ultrasound and 10 min of Ultra Turrax.

The benzene was eliminated by solidification of the mixture at  $0^\circ\text{C}$  and dynamic pumping of the vapors with the trapping of such at  $180^\circ\text{C}$ . After this last treatment, the flat micrometer-sized graphite was obtained in the dry state.

The distribution of the size of the particles subjected to stirring in a carrier liquid was determined using an apparatus of the Malvern type. Figures 2 and 3 respectively give the change in

the size of the particles as a function of the crushing time for a mixture containing 1 g/L and 10 g/L of graphite, respectively. One notes that all the particles have a diameter less than 30  $\mu\text{m}$ , and that 33% of the particles have a diameter between 4 and 14  $\mu\text{m}$ .

Figure 4a and Figure 4b respectively represent the grain size distribution histogram of the product obtained from a mixture containing 10 g/L crushed for 5 h and for 11 h.

The particles obtained have a thickness less than 0.05  $\mu\text{m}$  and a specific area on the order of 40  $\text{m}^2/\text{g}$ , and consequently a covering power of 20  $\text{m}^2/\text{g}$ .

The electron diffraction diagram of the flat micrometer-sized graphite obtained indicates a quasi-perfect monocrystallinity of the material.

The flat micrometer-sized graphite GMP of the present invention can be advantageously used as electrode material in rechargeable or nonrechargeable batteries. For this application, the flat micrometer-sized graphite is used in the form of a mixture with  $\text{MnO}_2$  or with any other material whose electrochemical and/or morphological properties are similar to those of  $\text{MnO}_2$ , for example,  $\text{V}_2\text{O}_5$ . In effect, a percolation phenomenon occurs in the GMP- $\text{MnO}_2$  mixtures, even in the case of GMP contents on the order of 1%. Example 6 in the following illustrates this property.

#### Example 6

A mixture of GMP and  $\text{MnO}_2$  was homogenized after having been wetted by cyclohexane. The homogenization was done using an ultrasound probe with a power of 500 W in a volume of 100  $\text{cm}^3$  with a frequency of 20 kHz for 10 min. The cyclohexane was then extracted from the mixture by lyophilization. The recovered powder

was made into pellets under a pressure of 1000 kg/cm<sup>2</sup>. The measurement of the electrical conductivity was done using the pellets obtained by the method of Van Der Pauw. According to this method, on the pellets mentioned above, contacts with silver lacquer as thin as possible at the four summits A, B, C, and D of a rectangle were produced. In a first step, a weak current on the order of a  $\mu\text{A}$ , or even a nA, was allowed to flow from A to B, noting the value of the voltage appearing between C and D for different intensities of the current. The curve representing the voltage as a function of the intensity of the current is a line with a slope R1. The operation was repeated, running a current from D to A, and the voltage appearing between points B and C was noted. The line representing the variation of the voltage between B and C as a function of the intensity of the current has a slope R2. The resistivity  $\rho$  of the layer formed by the pellets is given by the following equation:

$$\rho = \frac{\pi e}{2 \ln 2} (R1 + R2) f\left(\frac{R1}{R2}\right)$$

in which e represents the thickness of the layer, R1 and R2 repressed the resistances obtained by the measurements of voltage and current, and  $f(R1/R2)$  represents the tabulated function of Van Der Pauw.

The results of the measurements are collected in the following table.

Table

1 % volumique de GMP	2 Epaisseur de la pastille (en cm)	3 Résistivité ( $\Omega\text{cm}$ )
10	0,67	0,022
6	1,35	0,064
3	1,65	0,280
1	0,48	4,47



Key: 1      % by volume of GMP  
     2      Thickness of the pellet (in cm)  
     3      Resistivity ( $\Omega \cdot \text{cm}$ )

These results show that the phenomenon of percolation is attained in these two-phase media with a GMP content as low as 1%.

In the conventional batteries, particles of graphitized carbon black are spheroidal, and the percolation threshold is only obtained with percentages of carbon black in the vicinity of 30%.

All the properties of the particles of flat micrometer-sized graphite of the present invention allow them to be used for numerous applications. Thus, the particles of the present invention can be used in conventional tribology. They can also be used in the form of intercalated composites with an appropriate inserted element for applications of tribology under vacuum or at high temperature. They can also be used as they are, mixed with pitch, in order to promote graphitization of the pitch by a heat treatment.

### Claims

1. A graphitized material characterized by the fact that it essentially consists of monocrystalline particles with a form factor between 50 and 1000 and that, after deposition in the form of a layer a few millimeters thick on a flat support, have a mosaic structure, determined by X-ray diffraction, on the order of a  $\pm 15^\circ$  angle.

2. A graphitized material according to Claim 1, characterized by the fact that the form factor is greater than 100.

3. A process for the production of a graphitized material according to either Claim 1 or Claim 2, characterized by the fact that it consists of preparing an expanded graphite from a precursor material that is or can be graphitized, then crushing the expanded graphite obtained, with said crushing being done by the alternating action of an ultrasound probe and a rotating crusher with a shearing effect.

4. A process according to Claim 3, characterized by the fact that the precursor material is expanded by including an intercalated material in said substance, then by subjecting the intercalated composite obtained to a sudden temperature rise.

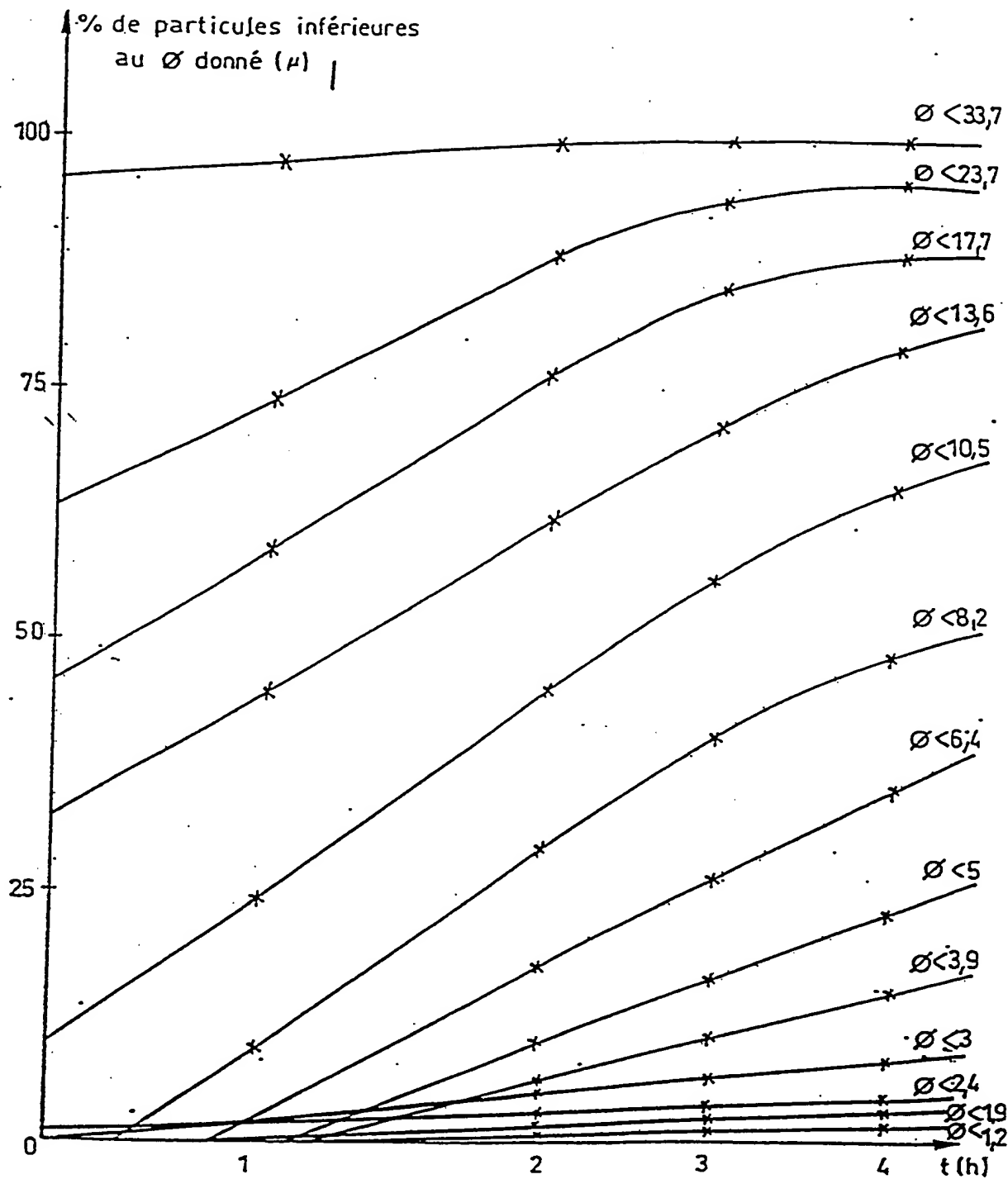
5. A process according to Claim 3, characterized by the fact that the expanded graphite is precrushed in a wetting liquid.

6. A process according to Claim 3, characterized by the fact that the wetting liquid is eliminated after crushing.

7. A process according to Claim 6, characterized by the fact that the wetting liquid is eliminated by lyophilization.

8. Application of the graphitized material according to either Claim 1 or Claim 2, for the preparation of electrode materials.

9. Application of the graphitized material according to either Claim 1 or Claim 2, for the graphitization of pitch.



Key: 1 % of particles less than the designated  $\phi$  ( $\mu$ )

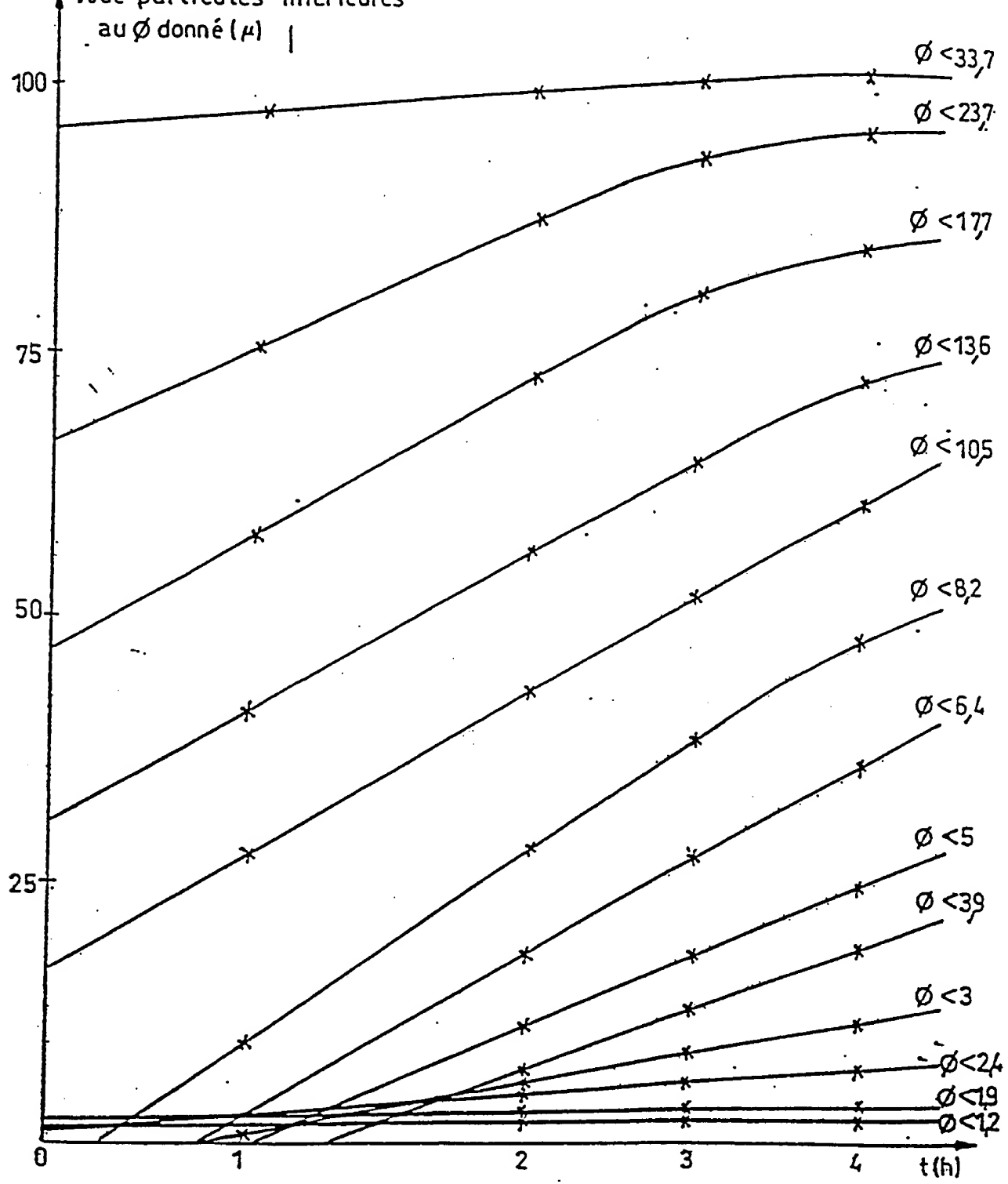


Figure 2

Key: 1 % of particles less than the designated  $\phi$  ( $\mu$ )

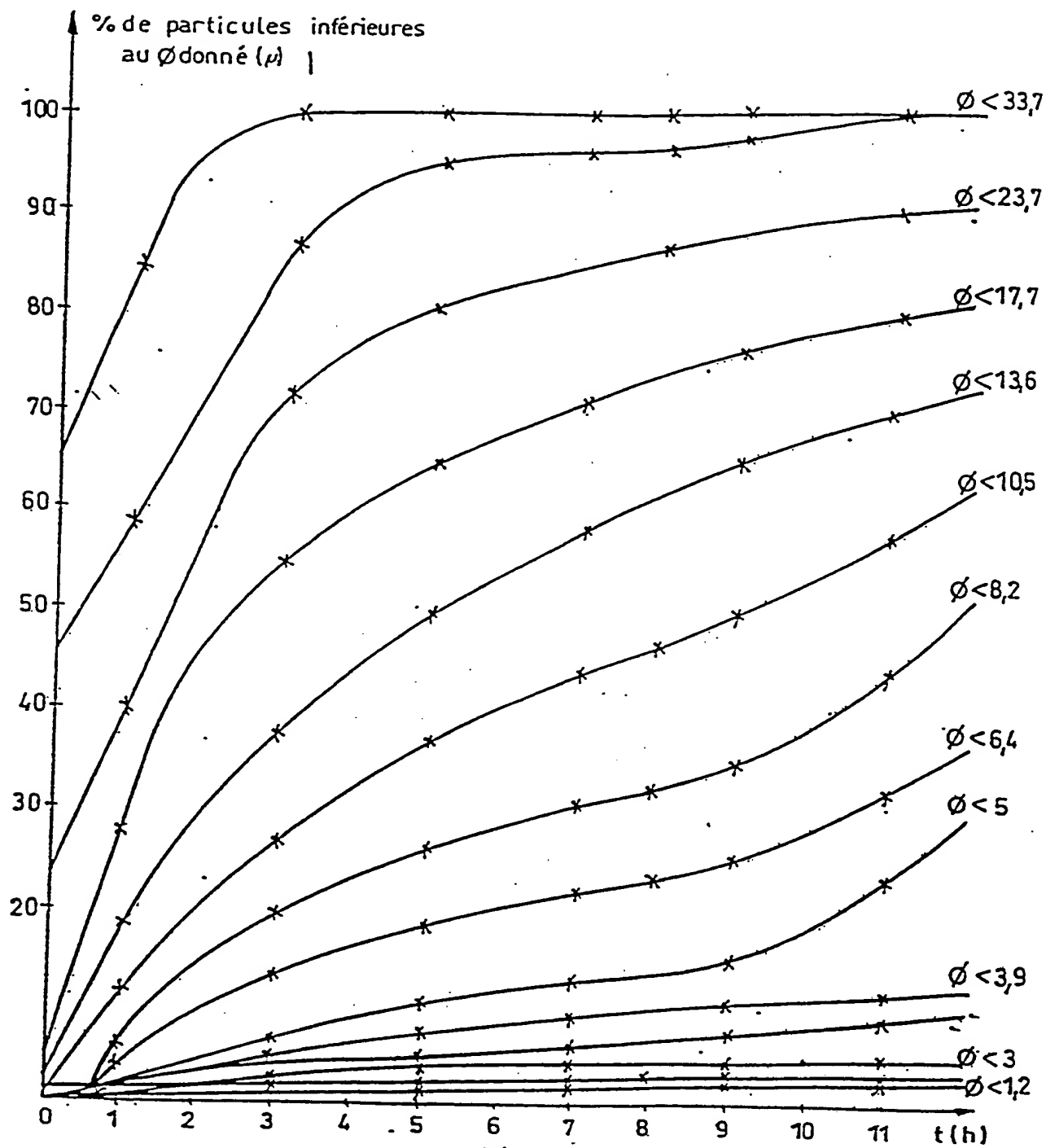


Figure 3

Key: 1    % of particles less than the designated  $\phi$  ( $\mu$ )

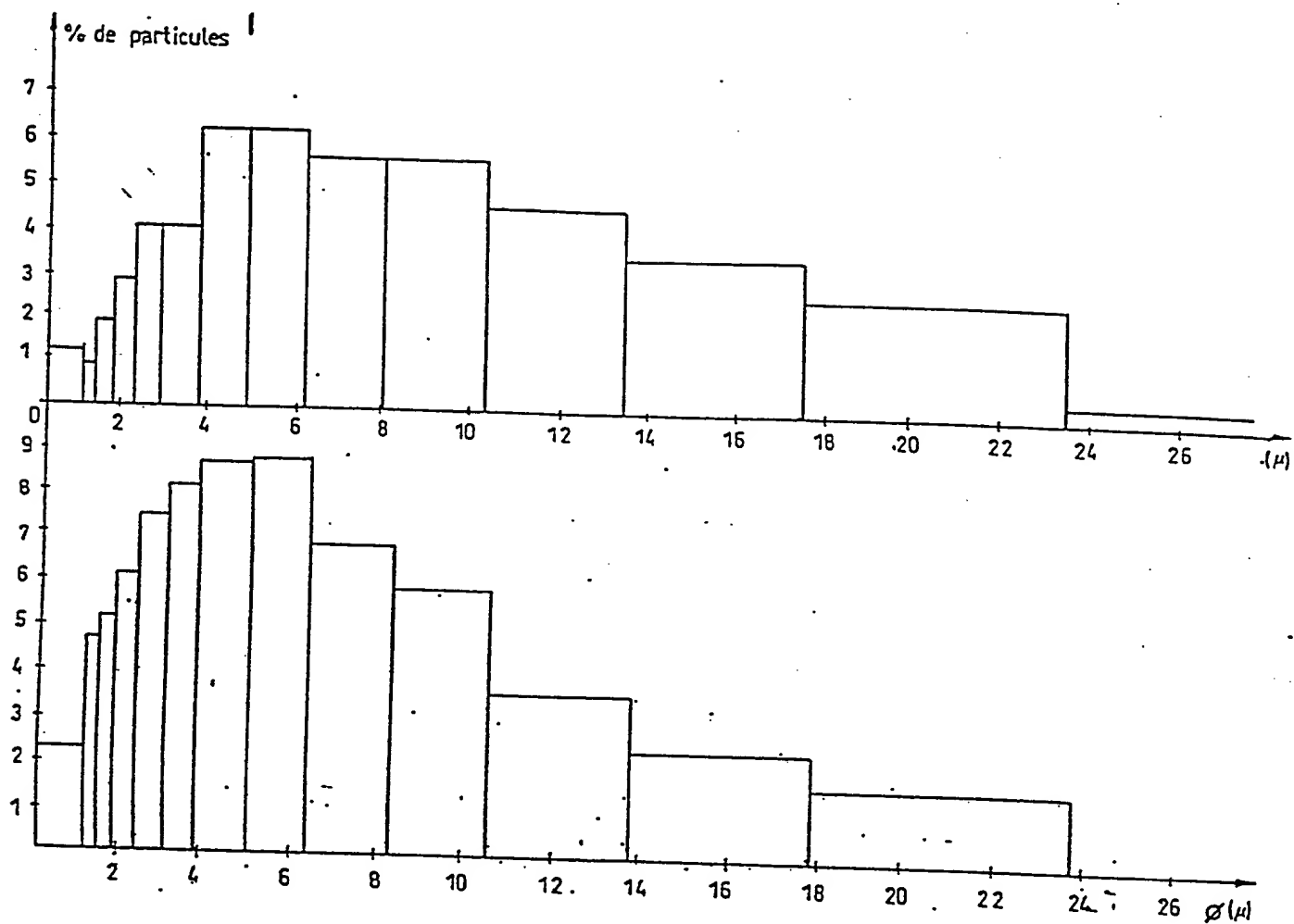


Figure 4

Key: 1      % of particles

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/EP 92/02317

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 5 C01B31/04

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 5 C01B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	CHEMICAL ABSTRACTS, vol. 100, no. 12 19 March 1984, Columbus, Ohio, US; abstract no. 91812p, CLARKE ET AL "A new adsorption substrate: Single crystal exfoliated graphite" page 384 ; & J. Appl. Phys. 1984, vol. 55, no. 4, pages 1231-3.	1
A	PATENT ABSTRACTS OF JAPAN vol. 10, no. 314 (C-380)(2370) 24 October 1986 & JP,A,61 127 612 ( AGENCY OF IND SCIENCE & TECHNOL ) 14 June 1986 -/-	1-3



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Date of the actual completion of the international search

22 December 1992 (22.12.92)

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## INTERNATIONAL SEARCH REPORT

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PCT/EP 92/02317

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	PATENT ABSTRACTS OF JAPAN vol. 12, no. 410 (C-540)(3257) 28 October 1988 & JP,A,63 147 810 ( HITACHI CHEM CO LTD ) 20 June 1988	3
A	PATENT ABSTRACTS OF JAPAN vol. 14, no. 406 (C-754)(4349) 4 September 1990 & JP,A,2 153 810 ( MITSUI MINING CO LTD ) 13 June 1990	3
A	DE,A, 3 505 656 (SIGRI GMBH) 28 August 1986 see claim 1	3



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ON INTERNATIONAL PATENT APPLICATION NO.

EP 9202317  
SA 65874

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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
DE-A-3505656	28-08-86	None	

International Patent No. WO 93/08123

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P.O. Box 4828, Austin, Texas 78765 USA